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13. ABSTRACT (Maximum 200 words) This program has supported students working on developing a fundamental understanding of the quantum features of nano-optical structures in semiconductors. A number of published papers resulted including a report on the first observation of the Raman coherence in a semiconductor, and the first report of directly probing optical excitation based on the coherent nonlinear optical response using a near-field optical microscope.			
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FINAL REPORT
To the Air Force Office of Scientific Research
AFOSR-AASERT
QUANTUM OPTICAL STUDIES OF SEMICONDUCTORS

AFOSR GRANT NO. F49620-96-1-0189

(PARENT AFOSR GRANT NO. F49620-96-1-0062)

(NOTE: Due to the parallel nature of the work on this program and the parent program, many of the results presented in this report are also presented in the report on the parent program)

GRANT PERIOD: 5/1/96 - 4/30/00

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Publication Record

Articles for Refereed Journals

- Anne C. Schaefer and Duncan G. Steel, "Nonlinear Optical Response of the GaAs Exciton-Polariton," *Physical Review Letters* **79**, 4870 (1997).
- K.B. Ferrio and D.G. Steel, "Excitonic Raman Quantum Beats in GaAs," *Physical Review Letters* **80**, 786 (1998).
- Michael Lewis, Peter Wolanin, Ari Gafni, Duncan Steel, "Near-Field Optical Microscopy of Single Molecules by Femtosecond Two-Photon Excitation," *Optics Letters* **23**, 1111-1113(1998).
- N. H. Bonadeo, A. S. Lenihan, Gang Chen, J. R. Guest, D. G. Steel D. Gammon, D. S. Katzer and D. Park, "Single Quantum Dot States Measured by Optical Modulation Spectroscopy, *Appl. Phys. Lett.* **75**, pp2933-2935 (1999).

Invited Papers

- Jeff Guest, D. Gammon, N.H. Bonadeo, J. Erland, D.G. Steel, E.S. Snow, D.S. Katzer and D. Park, "Localized Excitons: Probing One Quantum Dot at a Time" MRS-99.

Conference Presentations

- J. C. Kim, G. Chen, A. C. Schaefer, N. Bonadeo, D. Gammon and D. G. Steel, "Strongly Localized Excitons in a Narrow Single Quantum Well: Ensemble Dynamics of Single-Quantum-Dot Excitons," QELS'97 OSA Tech. Dig. **12**, pp 148-149 (1997).
- A. C. Schaefer, N. H. Bonadeo and D. G. Steel, "The Coherent Nonlinear Optical Response of the Exciton-Polariton," QELS'97 OSA Tech. Dig. **12**, pp 16-17 (1997).
- N. H. Bonadeo, D. Gammon and D. G. Steel, "Resonant Nonlinear Optical Response of a Single Quantum Dot," QELS'97 OSA Tech. Dig. **12**, pp 64-65 (1997).
- N. H. Bonadeo, A. S. Lenihan, D. Gammon and D. G. Steel, "Single quantum dot-like excitonic states measured by optical modulation spectroscopy," QELS'97 OSA Tech. Dig. **12**, pp 16-17 (1997).
- J.C. Kim, G. Chen, A.C. Schaefer, N.H. Bonadeo, D.G. Steel, D. Gammon, "Dynamics of strongly localized excitons," *Bulletin Am. Phys. Soc.* **42**, p70 (1997).
- N.H. Bonadeo, A.S. Lenihan, D.G. Steel, D. Gammon, "Nonlinear spectroscopy of single quantum dot states," *Bulletin Am. Phys. Soc.* **42**, p70 (1997).
- A.C. Schaefer, N.H. Bonadeo, D.G. Steel, "Nonlinear optical response of the exciton-polariton due to exciton-exciton interactions," *Bulletin Am. Phys. Soc.* **42**, p204 (1997).
- Gang Chen, Anne Schaefer, Dan Gammon, Duncan Steel, "Disorder Induced Interference in Exciton Decay Dynamics," IQEC'98 OSA Technical Digest **7**, p225-226 (1998).

- M.K. Lewis, P. Wolanin, A. Gafni, D.G. Steel, "Two-Photon Induced Fluorescence Imaging of Single Molecules Using Near-Field Scanning Optical Microscopy," IQEC '98, OSA Technical Digest 7 p227-228 (1998).
- J. R. Guest, T. H. Stievator, A. S. Lenihan, Gang Chen, D. Gammon,, D. S. Katzer, D. Park, D. G. Steel, "Nano-Optics: Imaging the Resonant Nonlinear Response of Individual Localized Excitons," QELS'99 (1999).
- J. R. Guest, T. H. Stievator, D. G. Steel Anthony Lenihan, "Nonlinear Near-Field Spectroscopy and Microscopy of Single Excitons in a Disordered Quantum Well," QELS'00
- A.S. Lenihan, M.V.G. Dutt, D.G. Steel, W. Schoenfeld, P.M. Petroff "Transient Optical Excitation and Control in Self-Assembled Quantum Dots," QELS'00.
- T. H. Stievator, A. S. Lenihan and D. G. Steel, D. Gammon, D. S. Katzer and D. Park, "Strong-Field Nonlinear Response of Quantum Dots," QELS'00.

Brief Review of Performance Highlights

The cited papers above provide a detailed report of the work performed on this AASERT and other details are contained in the Progress Reports. However, a few results have become very important and are summarized here.

Observation of the Oscillation of Raman Coherence

A key question in manybody physics in semiconductors has been the issue of the importance of higher-order Coulomb correlations that lead to biexcitons, for example. In this work, we designed an experiment to directly probe the nature of this interaction by looking for heavy-hole-light-hole (hh-lh) Raman coherence.

There have been numerous reports in the literature of so-called lh-hh quantum beats. However, these experiments are all based on four-wave mixing and in fact probe the lh-hh dipole coherence, not the Raman coherence. To detect the Raman which reports on the existence of higher Coulomb correlations (i.e., if there are higher order Coulomb correlations, there are no Raman coherence beats but there are dipole coherence beats), we developed a special experiment which measures the time evolution of the quantum phase. Theoretical predictions based on the master equations including the effects of higher order Coulomb correlations are shown in Fig. 1. Experimental results are shown in Fig. 2. The measurements are performed on a thin film of GaAs, but have important implications for our studies of nano-optics in semiconductors. Namely, the results demonstrate that we have developed a means to demonstrate the role of these higher order effects. In addition, the measurements show that we can detect the existence of entangled states (this work is supported on the parent grant, but not on the AASERT).

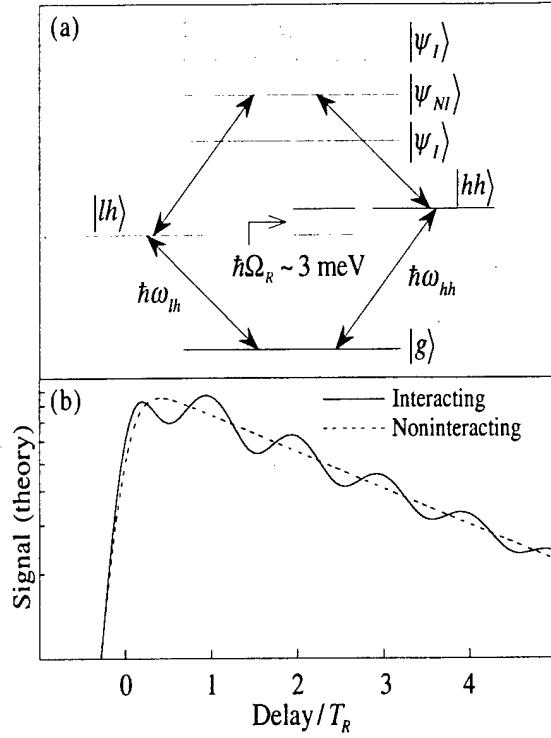


Figure 1. The lh - hh interaction produces bound or scattering states $|\psi_I\rangle$ (a) and produces Raman quantum beats (b: solid), which would be absent for noninteracting excitons (b: dashed). Energy differences have been greatly exaggerated for clarity. The Raman period and frequency are related by $T_R\Omega_R = 2\pi$.

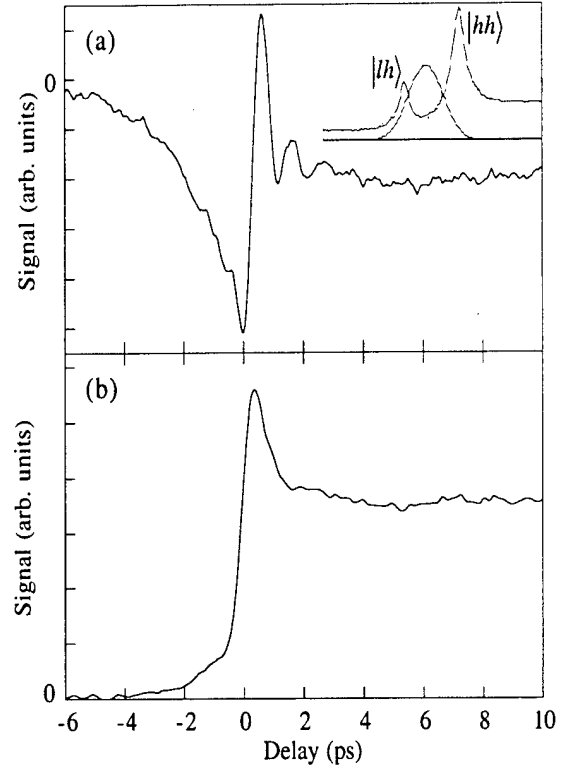


Figure 2. (a). Raman coherence oscillations appear in homodyne-detected FWM for the tuning shown in the inset. The pulse bandwidth is 2 meV, and the total excitation density is $9 \times 10^{15} \text{ cm}^{-3}$. (b). For a slightly different tuning, the beats are suppressed. (The overall sign of the DT response changes from (a) to (b), reflecting the sign change in EID as the laser is tuned closer to the lh exciton resonance.)

Development and Application of the First Low Temperature Near Field Scanning Microscope using High Resolution Coherent Nonlinear Optical Techniques: A Direct Optical Probe of the Local Density of States

A key aspect of the present program was development of low temperature near field optical microscope using the coherent nonlinear optical interaction as the probe rather than luminescence. This was essential to eliminate limitations that naturally accompany the use of fixed apertures or mesas, as well as enabling direct imaging of the localized states to better understand fundamental physical phenomena associated with disorder. The system will play a key role in the next generation of experiments.

The early work in near-field optical microscopy (NSOM) demonstrated the feasibility of obtaining *images* of electronic structures far beyond the classical limit by probing a system through a small aperture at the end of a scanning probe. This technology has been used to obtain photoluminescence (PL) from single molecules and semiconductor nanostructures. However, the current embodiment of these methodologies, however, relies on either the indirect optical probe of PL, which requires spectral and often spatial diffusion for detection, or sacrifices spatial information by probing the system through fixed apertures.

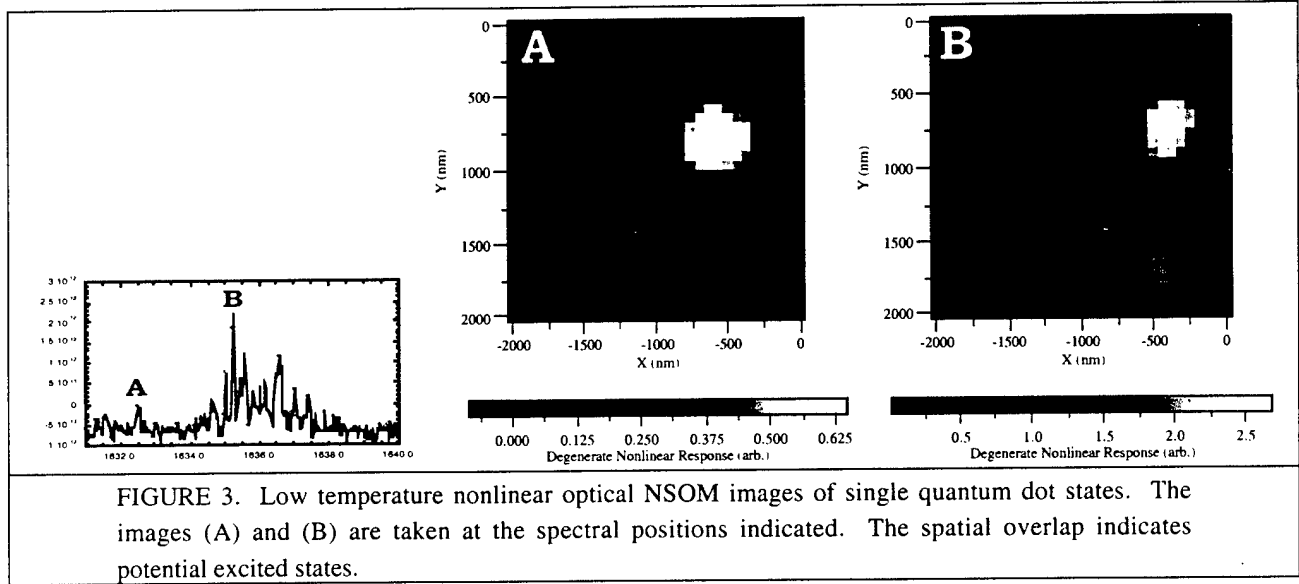
Here, we have developed a major advance in these technologies by combining the *direct* optical probe and spectral selectivity (at the nano-eV level) of coherent continuous-wave (CW) nonlinear spectroscopy with the spatial selectivity of near-field microscopy. This approach enables us to excite and probe the same quantum transition in an extended structure with the subwavelength resolution inherent in the NSOM methodology for the first time. At the most fundamental level, this capability allows us to map the optical dipole in space and energy with ultra-high resolution, revealing the local density of states (LDOS) of the system in analogy to previous scanning tunneling microscopy. This “quantum blueprint” of the eigenstates sets the stage for statistical analysis and allows for the identification of spectral and spatial correlations in the system.

The near-field microscope was constructed to operate at 4 Kelvin in order to reduce thermal broadening and perform high resolution spectroscopy on these exciton systems. The scanning probe was a single mode optical fiber that was ‘tube’ etched and coated with 100 nm of aluminum. The optical fields were coupled into this tip, which was brought into the vicinity of the sample (< 10 nm) and maintained there by feeding back on a shear force signal obtained through a phase-sensitive piezo-microphone.

The signal was collected in transmission, either with a photodiode located directly behind the sample stage or with collection optics and a multimode fiber to recover and direct the transmitted light to a spectrometer or an APD. Two interferometers were employed to verify the calibration and orthogonality of X and Y axes at 4 Kelvin. Images of the nonlinear response were obtained by raster scanning this high spatial resolution probe over the two dimensional quantum well with the lasers fixed at Ω_{probe} and Ω_{pump} .

Coherent CW nonlinear spectroscopy was implemented in our usual approach except that we had to work in a fully collinear geometry. The fields were linear and co-polarized at the sample. High resolution nonlinear spectra were obtained by positioning the microscope probe at a point in space and recording the signal as the lasers were tuned.

Direct evidence for localization can only be obtained by mapping the degenerate nonlinear response of an excitonic transition in space. Fig. 3 shows the nonlinear excitonic response of a $2\ \mu\text{m} \times 2\ \mu\text{m}$ region of the quantum well at two different energies, clearly displaying the localization and the isolation of the exciton eigenstate for the first time as showing the presence of an excited state.



In order to explore the CW nonlinear response thoroughly and make the connection with LDOS, it is necessary to explore the complete 3D data set. Fig. 4A shows the CW degenerate nonlinear response from a $7.44\ \text{meV} \times 2\ \mu\text{m} \times 2\ \mu\text{m}$ region. In this representation, eigenstates can be identified by their sharp horizontal line character. While most states congregate at the center of the quantum well in energy space, some states are scattered above and below the inhomogeneous line center. The low energy states have been shown to display quantum dot behavior in these systems. A set of states at higher energy, visible in the left-most panel and rendered in a volumetric representation in Fig. 4B, are localized to the same region of space and indicate that the local QW potential supports higher energy eigenstates. In addition, some of the higher energy states show some negative signal, indicating that the lines are being shifted or broadened by the application of the pump field, suggesting that higher order coulomb interactions are playing a role. It should be reiterated that these measurements are only sensitive to fast processes ($<1.6\ \text{ns}$), and there is no evidence for any phase shifts at 100 MHz.

By mapping the excitonic dipole directly in space and energy and therefore revealing the eigenstates of the system, it is clear that an analog to the LDOS is being uncovered. If the observed CW degenerate nonlinear response is the result of many isolated eigenstates, it should,

like the Schrödinger equation, be separable in energy and space: $S_{NL}(\hbar\Omega, \mathbf{R}) = \sum_n f_n(\hbar\Omega) h_n(\mathbf{R})$,

Where $f_n(\hbar\Omega)$ is the lineshape of nonlinear response of a single excitonic eigenstate and $h_n(\mathbf{R})$ is a spatial convolution of the excitonic response and the near-field probe response. $\hbar\Omega$ denotes the energy of the incident photons and \mathbf{R} is again the two-dimensional vector describing the lateral probe position on the sample. When saturation dominates the nonlinear response, as it does for these systems, the degenerate nonlinear response is simply the *saturation of the LDOS*.

With the identification and location of the eigenstates of the system, it is possible to look for spectral and spatial correlations between these states. For the disordered QW system studied in this work, Random Matrix Theory predicts that the eigenstates of the system will repel each other in energy and space. Spectrally, this behavior can be theoretically described by averaging many 2nd order correlation functions applied to high resolution spectra (dropping the \hbar): $g_2(\Delta\omega) \approx \left\langle \sum_{a \neq b} \delta(\omega_a - \omega_b - \Delta\omega) \right\rangle_R$. Experimentally, an analog to this expression is utilized in order to extract this information for the saturated LDOS: $g_2(\Delta\Omega) \approx \left\langle \int d\Omega S_{NL}(\Omega, \mathbf{R}) S_{NL}(\Omega - \Delta\Omega, \mathbf{R}) \right\rangle_R$. This function is plotted in Fig. 4C and shows a dip at 100 μeV which is the signature of spectral level repulsion.

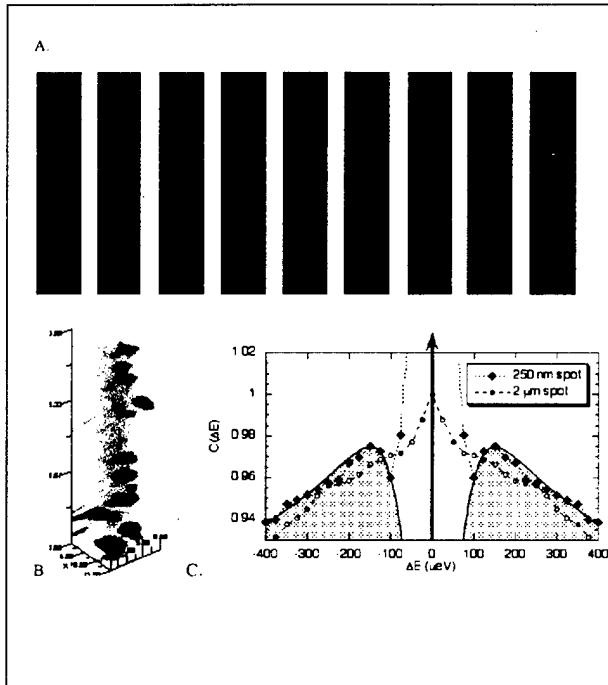


Fig. 4. 3D CW nonlinear response which represents $S(E, x, y)$. (A) The data set covers $2 \mu\text{m} \times 2 \mu\text{m} \times 7.44 \text{ meV}$. The vertical axis is energy (1588.44 meV to 1581.00 meV, top to bottom), the horizontal axis is x , and each 2D slice of data is taken at a different y value. (B) A volumetric representation of higher energy states in the left most panel of (A). (C) The correlation functions for 256 high spatial resolution spectra are calculated and averaged together (diamonds). They are compared with the correlation function for the spectrum obtained from a $2 \mu\text{m}$ spot (open circles), which is simulated by averaging the 256 high spatial resolution spectra. The correlation function for the high spatial resolution spectra shows a dip at 100 μeV which is consistent with the predictions of level repulsion. This dip is absent from the correlation function for the $2 \mu\text{m}$ spot spectrum. The shaded curves indicate the expected signature of $C(\Delta E)$ in the absence of a finite linewidth of the states, which dominates the measurement near $\Delta E=0$.

Summary

This AASERT has supported a number of students who have made major contributions to the parent program. The future work continues to emphasize nano-optics and quantum devices, and the progress made on this AASERT will be instrumental in our future successes.